

FORM PTO-1390 U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371		ATTORNEY'S DOCKET NUMBER: S 4731  U.S. APPLN. NO. (if known, see 37 CFR) <b>09/856903</b>
INTERNATIONAL APPLICATION NO.: PCT/FR99/02886	INTERNATIONAL FILING DATE: 23 NOVEMBER 1999	PRIORITY DATE CLAIMED: 27 NOVEMBER 1998
TITLE OF INVENTION: NOVEL ALKANOLAMIDE-FREE THICKENING LATEX		
APPLICANT(S) FOR DO/EO/US: Paul MALLO and Guy TABBACHI		
Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:		
1. <input checked="" type="checkbox"/> 2. <input type="checkbox"/> 3. <input checked="" type="checkbox"/> 4. <input checked="" type="checkbox"/> 5. <input checked="" type="checkbox"/> 6. <input checked="" type="checkbox"/> 7. <input type="checkbox"/> 8. <input type="checkbox"/> 9. <input type="checkbox"/> 10. <input type="checkbox"/> 11. <input checked="" type="checkbox"/> 12. <input type="checkbox"/> 13. <input checked="" type="checkbox"/> 14. <input type="checkbox"/> 15. <input type="checkbox"/> 16. <input checked="" type="checkbox"/>	This is a <b>FIRST</b> submission of items concerning a filing under 35 U.S.C. 371.  This is a <b>SECOND</b> or <b>SUBSEQUENT</b> submission of items concerning a filing under 35 U.S.C. 371.  This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).  A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.  A copy of the International Application as filed (35 U.S.C. 371(c)(2)) a. <input checked="" type="checkbox"/> is transmitted herewith (required only if not transmitted by the International Bureau). b. <input checked="" type="checkbox"/> has been transmitted by the International Bureau. (see attached copy of PCT/IB/308) c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US).  A translation of the International Application into English (35 U.S.C. 371(c)(2)).  Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3)). a. <input type="checkbox"/> are transmitted herewith (required only if not transmitted by the International Bureau). b. <input type="checkbox"/> have been transmitted by the International Bureau. c. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired. d. <input type="checkbox"/> have not been made and will not be made.  A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).  An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).  A translation of the annexes of the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).  Item 11. to 16. below concern document(s) or information included:  An Information Disclosure Statement under 37 CFR 1.97 and 1.98.  An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.  A <b>FIRST</b> preliminary amendment.  A <b>SECOND</b> or <b>SUBSEQUENT</b> preliminary amendment.  A substitute specification.  A change of power of attorney and/or address letter.  Other items or information:	
International Search Report PCT/IB/308 PCT/IPEA/409 Application Data Sheet		

U.S. APPLICATION NO. (USPTO form 37 CFR 1.51)

09/856903

INTERNATIONAL APPLICATION NO.  
PCT/FR99/02886ATTORNEY'S DOCKET NO.  
S 473117. ☒ The following fees are submitted:**BASIC NATIONAL FEE (37 CFR 1.492(a)(1)-(5)):**

Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO ..... \$ 1,000.00

International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO ..... \$ 860.00

International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO ..... \$ 710.00

International preliminary examination fee (37 CFR 1.482) paid to USPTO but all claims did not satisfy provisions of PCT Article 33(1)-(4) ..... \$ 690.00

International preliminary examination fee (37 CFR 1.482) paid to USPTO and all claims satisfied provisions of PCT Article 33(1)-(4) ..... \$ 100.00

**ENTER APPROPRIATE BASIC FEE AMOUNT =****CALCULATIONS PTO USE ONLY**

\$ 860.00

Surcharge of \$130.00 for furnishing the oath or declaration later than 30 months from the earliest claimed priority date (37 CFR 1.492(e)).

\$ 130.00

CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE	\$
Total claims	12 - 20 =	0	X \$18.00	\$
Independent claims	1 - 3 =	0	X \$80.00	\$
MULTIPLE DEPENDENT CLAIMS(S) (if applicable)			+ \$270.00	\$

**TOTAL OF ABOVE CALCULATIONS =**

\$ 990.00

Reduction of 1/2 for filing by small entity, if applicable. Applicant claims Small Entity Status under 37 CFR 1.27.

+

**SUBTOTAL =**

\$ 990.00

Processing fee of \$130 for furnishing the English translation later than months from the earliest claimed priority date (37 CFR 1.49(f)).

\$

**TOTAL NATIONAL FEE =**

\$ 990.00

Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property +

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**TOTAL FEES ENCLOSED =**

\$ 990.00

Amount to be  
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a. ☒ A check in the amount of \$990.00 to cover the above fees is enclosed.b. ☐ Please charge my Deposit Account No. **25-0120** in the amount of \$ to cover the above fees. A duplicate copy of this sheet is enclosed.c. ☒ The Commissioner is hereby authorized to charge any additional fees which may be required by 37 CFR 1.16 and 1.17, or credit any overpayment to Deposit Account No. **25-0120**. A duplicate copy of this sheet is enclosed.

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May 29, 2001

By

  
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09/856903

JC18 Rec'd PCT/PTC 29 MAY 2001

PATENTS

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of

Paul MALLO et al.

Serial No. (unknown)

Filed herewith

NOVEL ALKANOLAMIDE-FREE  
THICKENING LATEX

PRELIMINARY AMENDMENT

Commissioner for Patents

Washington, D.C. 20231

Sir:

Prior to the first Official Action and calculation of the filing fee, please substitute Claims 1-13 as originally filed, with Claims 1-12 as filed in the Article 34 amendment of December 8, 2000. The pages containing Claims 1-12 are marked "AMENDED SHEET" and are attached hereto. Following the insertion of Claims 1-12, please amend these claims as follows:

IN THE CLAIMS:

Amend claim 3 as follows:

--3. (Amended) Process as defined in claim 1, in which the polymerization reaction is initiated with a redox couple which generates hydrogen sulfite ions ( $\text{HSO}_3^-$ ), such as the cumene hydroperoxide/sodium metabisulfite ( $\text{Na}_2\text{S}_2\text{O}_5$ ) couple or the cumene hydroperoxide/thionyl chloride ( $\text{SOCl}_2$ ) couple, at a temperature of less than or equal to  $10^\circ\text{C}$ , if desired, supplemented with a polymerization coinitiator, such as azobis (isobutyronitrile) (AIBN).

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Amend claim 4 as follows:

--4. (Amended) Process as defined in claim 1, characterized in that the anionic polyelectrolyte comprises from 30% to 50% of a monomer comprising a strong acid function and from 70% to 50% either of a monomer comprising a weak acid function or of a neutral monomer.

Amend claim 6 as follows:

--6. (Amended) Process as defined in claim 1, characterized in that the anionic polyelectrolyte is vulcanized and/or branched with a diethylenic or polyethylenic compound in a molar proportion, expressed relative to the monomers used, of from 0.005% to 1% and preferably from 0.01 to 0.1%.

Amend claim 8 as follows:

--8. (Amended) Process as defined in claim 1, characterized in that the oil phase represents from 15% to 40% and preferably from 20% to 25% of its total weight.

Amend claim 9 as follows:

--9. (Amended) Process as defined in claim 1, characterized in that the oil phase consists essentially of isohexadecane or of white mineral oil.

Amend claim 10 as follows:

--10. (Amended) Use of composition obtained according to the process as defined in claim 1, to prepare a cosmetic, dermopharmaceutical or pharmaceutical topical composition.

Amend claim 11 as follows:

--11. (Amended) Cosmetic, dermopharmaceutical or pharmaceutical composition comprising from 0.1% to 10% by weight of an inverse latex obtained according to the process as defined in claim 1.--

R E M A R K S

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The above changes in the claims merely place this national phase application in the same condition as it was during Chapter II of the international phase, with the multiple dependencies being removed. Following entry of this amendment by substitution of the pages, only claims 1-12 remain pending in this application.

Attached hereto is a marked-up version of the changes made to the claims by the current amendment. The attached page is captioned "VERSION WITH MARKINGS TO SHOW CHANGES MADE".

Respectfully submitted,

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By



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May 29, 2001

VERSION WITH MARKINGS TO SHOW CHANGES MADE

The claims have been amended as follows:

3. (Amended) Process as defined in ~~either of Claims 1 and 2~~, in which the polymerization reaction is initiated with a redox couple which generates hydrogen sulfite ions ( $\text{HSO}_3^-$ ), such as the cumene hydroperoxide/sodium metabisulfite ( $\text{Na}_2\text{S}_2\text{O}_5$ ) couple or the cumene hydroperoxide/thionyl chloride ( $\text{SOCl}_2$ ) couple, at a temperature of less than or equal to  $10^\circ\text{C}$ , if desired, supplemented with a polymerization coinitiator, such as azobis (isobutyronitrile) (AIBN).

4. (Amended) Process as defined in ~~one of Claims 1 to 3~~, characterized in that the anionic polyelectrolyte comprises from 30% to 50% of a monomer comprising a strong acid function and from 70% to 50% either of a monomer comprising a weak acid function or of a neutral monomer.

6. (Amended) Process as defined in ~~one of Claims 1 to 5~~, characterized in that the anionic polyelectrolyte is vulcanized and/or branched with a diethylenic or polyethylenic compound in a molar proportion, expressed relative to the monomers used, of from 0.005% to 1% and preferably from 0.01 to 0.1%.

8. (Amended) Process as defined in ~~one of Claims 1 to 7~~, characterized in that the oil phase represents from 15% to 40% and preferably from 20% to 25% of its total weight.

9. (Amended) Process as defined in ~~one of Claims 1 to 8~~, characterized in that the oil phase consists essentially of isohexadecane or of white mineral oil.

10. (Amended) Use of composition obtained according to the process as defined in ~~one of Claims 1 to 9~~, to prepare a cosmetic, dermopharmaceutical or pharmaceutical topical composition.

11. (Amended) Cosmetic, dermopharmaceutical or pharmaceutical composition comprising from 0.1% to 10% by weight

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of an inverse latex obtained according to the process as defined  
in ~~one of~~ claims ~~1 to 9~~.

TECHNICAL COOPERATION

**Novel alkanolamide-free thickening latex**

5 The present application relates to a novel process for water-in-oil latices, and to the application of these latices as thickeners and/or emulsifiers for skincare products and haircare products or for the manufacture of cosmetic, dermatopharmaceutical or pharmaceutical preparations.

10 Among the various existing thickeners which are used for these purposes, there are, in particular, synthetic thickening polymers, which are in the form of inverse latices, i.e. latices in which the continuous phase is an oil. These latices dissolve extremely quickly; the polymers contained in these inverse  
15 latices are, for example, acrylamide/sodium 2-acrylamido-2-methylpropanesulfonate copolymers; they are already neutralized and when they are dissolved in water, for example to a concentration of 1%, it is observed that the pH is generally greater than 6. Such  
20 inverse latices are disclosed in the European patent applications published under Nos. EP 0 186 361 and EP 0 503 853. These latices retain considerable thickening capacity even at pH 4.

25 However, the process for preparing such copolymers uses surfactants of the alkanolamide family, for instance Witcamide<sup>TM</sup> 511; now, these chemical compounds are liable to degrade into compounds of the nitrosamine family which are themselves products that are known as being potentially carcinogenic. The simple  
30 implementation of the precautionary principle thus makes it unavoidable that, in the short or long term, the inverse latices mentioned above will be banned from use in the preparation of cosmetic, dermatopharmaceutical or pharmaceutical products. Consequently, the Applicant  
35 has become interested in developing a novel process for synthesizing inverse latices which does not have this drawback.

One subject of the invention is a process for preparing a composition in the form of an inverse

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latex, comprising an oil phase, an aqueous phase, at least one emulsifier of water-in-oil (W/O) type, at least one emulsifier of oil-in-water (O/W) type and from 20% to 75% by weight, mainly from 20% to 60% by weight and more particularly from 30% to 45% by weight, of a branched or vulcanized anionic polyelectrolyte, this process successively comprising:

- a step (a) of preparing an aqueous solution containing the monomers and the optional additives;

- a step (b) of emulsifying the aqueous phase prepared in step (a), in an organic phase, in the presence of one or more emulsifiers of water-in-oil type;

- a step (c) of polymerizing the monomers in the aqueous phase, initiated by introducing a free-radical initiator into said phase; and

- a step (d) of adding one or more emulsifiers of oil-in-water type to the resulting dispersion, at a temperature of less than 50°C,

characterized in that:

- the polymerization reaction in step (c) is carried out at a pH of less than 5.5,

- none of said emulsifiers belongs to the alkanolamide family, and

- said anionic polyelectrolyte is based either on a monomer containing a strong acid function, or on at least one monomer containing a strong acid function copolymerized either with at least one monomer containing a weak acid function or with at least one neutral monomer.

According to one variant of this process, the reaction medium obtained from step (b) is concentrated by distillation before carrying out step (c).

According to one preferred embodiment of the process as defined above, the polymerization reaction is initiated with a redox couple which generates hydrogen sulfite ions ( $\text{HSO}_3^-$ ), such as the cumene hydroperoxide/sodium metabisulfite ( $\text{Na}_2\text{S}_2\text{O}_5$ ) couple, or the cumene hydroperoxide/thionyl chloride ( $\text{SOCl}_2$ )

couple, at a temperature of less than or equal to 10°C, if desired, supplemented with a polymerization coinitiator such as, for example, azobis(isobutyronitrile) (AIBN), and is then carried out either  
5 virtually adiabatically up to a temperature of greater than or equal to 50°C, or by controlling the temperature.

The term "alkanolamide" denotes the products resulting from the action of fatty acids in equimolar  
10 amounts on alkanolamines such as, for example, mono- or diethanolamine.

The expression "emulsifier of the water-in-oil type" denotes emulsifiers having an HLB value which is low enough to give water-in-oil emulsions such as the  
15 surfactant polymers sold under the name Hypermer™ or such as sorbitan extracts, for instance the sorbitan monooleate sold by the company SEPPIC under the brand name Montane™ 80, or the sorbitan isostearate sold by SEPPIC under the name Montane™ 70®.

The expression "emulsifier of the oil-in-water type" denotes emulsifiers with an HLB value which is high enough to give oil-in-water emulsions such as  
20 ethoxylated sorbitan esters, for instance sorbitan oleate ethoxylated with 20 mol of ethylene oxide.

The term "branched polymer" denotes a nonlinear polymer containing pendent chains so as to obtain, when this polymer is dissolved in water, a highly entangled  
25 state leading to very high viscosities at low shear.

The term "vulcanized polymer" denotes a  
30 nonlinear polymer in the form of a three-dimensional network which is insoluble in water, but swellable in water and thus leading to the production of a chemical gel. The composition obtained according to the process as defined above may comprise vulcanized moieties  
35 and/or branched moieties.

A subject of the invention is, more particularly, a process as defined above, characterized in that 30% to 80% and preferably 30% to 60%, in molar proportions, of the monomer moieties which the anionic

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polyelectrolyte comprises contain a strong acid function; and more particularly characterized in that the anionic polyelectrolyte comprises from 30% to 50% of a monomer comprising a strong acid function and from 5 70% to 50% either of a monomer comprising a weak acid function or of a neutral monomer.

The strong acid function in the monomer comprising it is, in particular, the sulfonic acid function or the phosphonic acid function, and said 10 monomer is preferably 2-methyl-2-[(1-oxo-2-propenyl)-amino]-1-propanesulfonic acid. or the acid. The weak acid function in the monomer comprising it is, in particular, the carboxylic acid function, and said monomer is preferably chosen from acrylic acid, 15 methacrylic acid, itaconic acid and maleic acid. The neutral monomer is chosen in particular from acrylamide, 2-hydroxyethyl acrylate, 2,3-dihydroxyethyl acrylate, 2-hydroxyethyl methacrylate, 2,3-dihydroxyethyl methacrylate, or an ethoxylated derivative, with 20 an EO number of between 1 and 20, for each of these esters.

A subject of the invention is, more particularly, a process as defined above, characterized in that the anionic polyelectrolyte comprises, in molar 25 proportions, from 30% to 50% of 2-methyl-2-[(1-oxo-2-propenyl)amino]-1-propanesulfonic [lacuna], partially or totally salified in the form of an alkali metal salt, preferably the sodium salt, or in the form of the ammonium salt, and from 70% to 50% of acrylamide.

30 A subject of the invention is, more particularly, a process as defined above, characterized in that the anionic polyelectrolyte is vulcanized and/or branched with a diethylenic or polyethylenic compound in a molar proportion, expressed relative to 35 the monomers used, of from 0.005% to 1% and preferably from 0.01% to 0.1%, and the vulcanizing agent and/or branching agent is chosen in particular from ethylene glycol methacrylate, sodium diallyloxyacetate, ethylene glycol diacrylate, diallylurea, trimethylolpropane

triacrylate and, more particularly, methylene-bis(acrylamide).

The latex obtained by the process according to the invention generally contains from 2.5% to 15% by weight and preferably from 4% to 9% by weight of emulsifiers, among which from 20% to 50% and in particular from 25% to 40% of the total weight of the emulsifiers present are of the water-in-oil (W/O) type and in which from 80% to 50% and in particular from 75% to 60% of the total weight of the emulsifiers are of the oil-in-water (O/W) type.

According to one particular aspect of the process as defined above, the emulsifiers of the water-in-oil type consist essentially of sorbitan monooleate.

According to another particular aspect, the composition obtained by the process as defined above is characterized in that the oil phase represents from 15% to 40% and preferably from 20% to 25% of its total weight. This oil phase consists either of a commercial mineral oil containing saturated hydrocarbons of paraffinic, isoparaffinic or cycloparaffinic type, having at room temperature a density of between 0.7 and 0.9 and a boiling point of greater than 180°C, such as, for example, Exxsol D 100 S sold by EXXON or a white mineral oil, such as Marcol<sup>TM</sup> 52, or the isohexadecane sold by Bayer or isododecane, or of a plant oil or of a synthetic oil, or of a mixture of several of these oils.

According to one preferred aspect of the present invention, the composition obtained by the process as defined above is characterized in that the oil phase consists essentially of isohexadecane or of Marcol<sup>TM</sup> 52. Isohexadecane, which is identified in Chemical Abstracts by the RN number = 93685-80-4, is a mixture of C<sub>12</sub>, C<sub>16</sub> and C<sub>20</sub> isoparaffins containing at least 97% C<sub>16</sub> isoparaffins, among which the main constituent is 2,2,4,4,6,8,8-heptamethylnonane (RN = 4390-04-9). Marcol<sup>TM</sup> 52 is a commercial oil corresponding to the definition of liquid petroleum of

the French Codex. This is a white mineral oil in accordance with FDA regulations 21 CFR 172.878 and CFR 178.3620(a) and is registered in the US Pharmacopoeia in US XXIII (1995) and in the European pharmacopoeia (1993).

The latices contain between 20% and 50% water. The latices according to the invention may also contain various additives such as complexing agents, transfer agents or chain-limiting agents.

A subject of the invention is also the use of the composition obtained according to the process as defined above, to prepare a cosmetic, dermo-pharmaceutical or pharmaceutical topical composition.

A topical composition according to the invention, intended to be applied to human or animal skin or mucous membranes, may consist of a topical emulsion comprising at least one aqueous phase and at least one oil phase. This topical emulsion may be of the oil-in-water type. More particularly, this topical emulsion may consist of a fluid emulsion, such as a milk or a fluid gel. The oil phase of the topical emulsion may consist of a mixture of one or more oils.

A topical emulsion according to the invention may be intended for cosmetic use or may be used to prepare a medicinal product intended for treating diseases of the skin and of mucous membranes. In the latter case, the topical composition then comprises an active principle which may consist, for example, of an anti-inflammatory agent, a muscular relaxant, an anti-fungal agent or an antibacterial agent.

When the topical composition is used as a cosmetic composition intended to be applied to the skin or mucous membranes, it may or may not comprise an active principle, for example a moisturizer, a tanning agent, a sunscreen, an anti-wrinkle agent, a slimming agent, a free-radical scavenger, an antiacne agent or an antifungal agent.

A topical composition according to the invention usually comprises between 0.1% and 10% by

weight of the thickener defined above. The pH of the topical composition is preferably greater than or equal to 5 and is more preferably between 6 and 12.

The topical composition may also comprise compounds conventionally included in compositions of this type, for example fragrances, preserving agents, colorants, emollients or surfactants.

According to yet another aspect, the invention relates to the use of the novel thickener in accordance with the invention mentioned above, to thicken and emulsify a topical composition comprising at least one aqueous phase.

The cosmetic, dermopharmaceutical or pharmaceutical composition defined above generally comprises from 0.1% to 10% and more particularly between 0.5% and 5% by weight of said inverse latex. It is in particular in the form of a milk, a lotion, a gel, a cream, a cream-gel, a soap, a bubble bath, a balm, a shampoo or a conditioner.

In general, said inverse latex may advantageously replace the products sold under the name Sepigel™ 305 or Sepigel™ 501 by the Applicant, in cosmetic, dermopharmaceutical or pharmaceutical compositions, since it also has good compatibility with the other excipients used for the preparation of formulations such as milks, lotions, creams, soaps, [lacuna] baths, balms, shampoos and conditioners. It may also be used in combination with said Sepigel products.

It is especially compatible with the concentrates disclosed and claimed in international publications WO 92/06778, WO 95/04592, WO 95/13863, WO 98/47610 and FR 2 734 496, or with the surfactants disclosed in WO 93/08204.

It is particularly compatible with Montanov™ 68, Montanov™ 82, Montanov™ 202 and Sepiperl™ N. It may also be used in emulsions of the type disclosed and claimed in EP 0 629 396 and in cosmetically or physiologically acceptable aqueous

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dispersions with a polyorganosiloxane compound chosen, for example, from those disclosed in WO 93/05762 or in WO 93/21316. It may also be used to form cosmetically or physiologically acceptable aqueous gels of acidic pH, such as those disclosed in WO 93/07856; it may also be used in combination with nonionic celluloses to form, for example, styling gels, such as those disclosed in EP 0 684 024, or alternatively in combination with fatty acid esters of sugars, to form compositions for treating the hair or the skin such as those disclosed in EP 0 603 019, or alternatively in shampoos or conditioners as disclosed and claimed in WO 92/21316, or finally in combination with an anionic homopolymer such as Carbopol<sup>TM</sup> to form hair treatment products such as those disclosed in DE 195 23596. It is also compatible with many active principles such as, for example, self-tanning agents, for instance dihydroxyacetone (DHA) or antiacne agents; it may thus be introduced into self-tanning compositions such as those claimed in EP 0 715 845, EP 0 604 249, EP 0 576 188 or WO 93/07902. It is also compatible with N-acyl amino acid derivatives, which allows its use in soothing compositions in particular for sensitive skin, such as those disclosed or claimed in WO 92/21318, WO 94/27561 or WO 98/09611. It is also compatible with glycolic acids, with lactic acid, with salicylic acid, retinoids, phenoxyethanol, sugars, glyceraldehyde, xanthans, fruit acids and the various polyols used in the manufacture of cosmetic formulations.

A subject of the invention is thus also the use of an inverse latex as defined above to prepare a cosmetic, dermatopharmaceutical or pharmaceutical composition.

The examples which follow are intended to illustrate the present invention without, however, limiting it.

**Example 1: Preparation of the latex according to the invention**

a) The following are loaded into a beaker, with stirring

- 5 - 80 g of deionized water,
  - 95.96 g of aqueous 48% (by weight) sodium hydroxide solution,
  - 246.7 g of 2-methyl-2-[(1-oxo-2-propenyl)amino]-1-propanesulfonic acid,
  - 10 - 253.8 g of 50% acrylamide,
  - 0.45 g of sodium diethylenetriaminepentaacetate,
  - 0.132 g of methylenebis(acrylamide);
- the pH of the aqueous phase described above is adjusted to about 5.0 and the amount of aqueous phase is made up
- 15 to 682 g by adding deionized water. In parallel, an organic phase is prepared by introducing the following successively into a stirred beaker:
  - 220 g of isohexadecane,
  - 21 g of Montane<sup>TM</sup> 80 VG (sorbitan oleate sold by
  - 20 SEPPIC)
  - 0.2 g of AIBN.

The aqueous phase is gradually introduced into the organic phase and is then subjected to vigorous mechanical stirring using an Ultra-Turrax<sup>®</sup> mixer sold

25 by IKA.

The emulsion obtained is then transferred into a polymerization reactor. The emulsion is bubbled with a strong stream of nitrogen so as to remove the oxygen, and is cooled to about 5-6°C.

30 5 ml of a solution containing 0.28% (by weight) of cumene hydroperoxide in isohexadecane are then added.

After a sufficient amount of time for good homogenization of the solution, an aqueous sodium

35 metabisulfite solution (2.5 g in 100 ml of water) is then added at a rate of 0.5 ml/minute. The addition is carried out over about 60 minutes. During this addition, the temperature in the polymerization reactor



is allowed to rise up to the final polymerization temperature.

The reaction medium is then maintained at this temperature for about 90 minutes.

5           The mixture is cooled to a temperature of about 35°C and 50 g of sorbitan oleate ethoxylated with 20 mol of ethylene oxide are added slowly.

The desired emulsion is obtained:

Evaluation of the properties

10 + viscosity in water containing 2% latex (Brookfield RVT spindle 6, speed 20): 23 450 mPa·s

(Brookfield spindle 6, speed 5):  $\eta$  = 69 000 mPa·s.

15           b) The process described above in paragraph a) is repeated, replacing the isohexadecane with Marcol™ 52, to prepare a latex based on white mineral oil.

The examples which follow use any one of the emulsions prepared in Example 1.

20   **Example 2: Care cream**

	Cyclomethicone:	10%
	Composition 1:	0.8%
	Montanov™ 68:	2%
	Stearyl alcohol:	1%
25	Stearic alcohol:	0.5%
	Preserving agent:	0.65%
	Lysine:	0.025%
	EDTA (disodium salt):	0.05%
	Xanthan gum:	0.2%
30	Glycerol:	3%
	Water:	qs 100%

**Example 3: Care cream**

	Cyclomethicone:	10%
35	Composition 1:	0.8%
	Montanov™ 68:	2%
	Perfluoropolymethyl isopropyl ether:	0.5%
	Stearyl alcohol:	1%

	Stearic alcohol:	0.5%
	Preserving agent:	0.65%
	Lysine:	0.025%
	EDTA (disodium salt):	0.05%
5	Pemulen™ TR:	0.2%
	Glycerol:	3%
	Water:	qs 100%

**Example 4: Aftershave balm**

10 FORMULA

A	Composition 1:	1.5%
	Water:	qs 100%
B	Micropearl™ M 100:	5.0%
	Sepicide™ CI:	0.50%
15	Fragrance:	0.20%
	95° ethanol:	10.0%

PROCEDURE

Add B to A.

20 **Example 5: Satin body emulsion**

FORMULA

A	Simulsol™ 165:	5.0%
	Lanol™ 1688:	8.50%
	Karite butter:	2%
25	Liquid paraffin:	6.5%
	Lanol™ 14M:	3%
	Lanol™ S:	0.6%
B	Water:	66.2%
C	Micropearl™ M 100:	5%
30	D Composition 1:	3%
E	Sepicide™ CI:	0.3%
	Sepicide™ HB:	0.5%
	Monteine™ CA:	1%
	Fragrance:	0.20%
35	Vitamin E acetate	0.20%
	Sodium pyrrolidinone- carboxylate:	1% (moisturizer)

PROCEDURE

Add C to B, emulsify B in A at 70°C, then add D at 60°C, followed by E at 30°C.

5 **Example 6: Body milk**

FORMULA

10	A	Simulsol™ 165:	5.0%
		Lanol™ 1688:	12.0%
		Lanol™ 14M:	2.0%
		Cetyl alcohol:	0.3%
		Schercemol™ OP:	3%
	B	Water:	qs 100%
	C	Composition 1:	0.35%
	D	Sepicide™ CI:	0.2%
15		Sepicide™ HB:	0.5%
		Fragrance:	0.20%

PROCEDURE

Emulsify B in A at about 75°C; add C at about 60°C, followed by D at about 30°C.

20

**Example 7: O/W cream**

FORMULA

25	A	Simulsol™ 165:	5.0%
		Lanol™ 1688:	20.0%
		Lanol™ P:	1.0% (additive with stabilizing effect)
	B	Water:	qs 100%
	C	Composition 1:	2.50%
30	D	Sepicide™ CI:	0.20%
		Sepicide™ HB:	0.30%

PROCEDURE

Introduce B into A at about 75°C; add C at about 60°C, followed by D at about 45°C.

35

**Example 8: Nongreasy antisun gel**

FORMULA

A	Composition 1:	3.00%
	Water:	30%

	B	Sepicide™ CI:	0.20%
		Sepicide™ HB:	0.30%
		Fragrance:	0.10%
5	C	Colorant:	qs
		Water:	30%
	D	Micropearl™ M 100:	3.00%
		Water:	qs 100%
	E	Silicone oil:	2.0%
		Parsol™ MCX:	5.00%
10	<u>PROCEDURE</u>		
	Introduce B into A; add C, then D, followed by E.		

**Example 9: Antisun milk**

FORMULA

15	A	Sepiperl™ N:	3.0%
		Sesame oil:	5.0%
		Parsol™ MCX:	5.0%
		λ carrageenan:	0.10%
	B	Water:	qs 100%
20	C	Composition 1:	0.80%
	D	Fragrance:	qs
		Preserving agent:	qs

PROCEDURE

Emulsify B in A at 75°C and then add C at about 60°C,  
25 followed by D at about 30°C, and adjust the pH if  
necessary.

**Example 10: Massage gel**

FORMULA

30	A	Composition 1:	3.5%
		Water:	20.0%
	B	Colorant:	2 drops/100 g
		Water:	qs
35	C	Alcohol:	10%
		Menthol:	0.10%
	D	Silicone oil:	5.0%

PROCEDURE

Add B to A; then add C to the mixture, followed by D.

**Example 11: Massage care gel**

FORMULA

	A	Composition 1:	3.00%
		Water:	30%
5	B	Sepicide™ CI:	0.20%
		Sepicide™ HB:	0.30%
		Fragrance:	0.05%
	C	Colorant:	qs
		Water:	qs 100%
10	D	Micropearl™ SQL:	5.00%
		Lanol™ 1688:	2%

PROCEDURE

Prepare A; add B and then C, followed by D.

15 **Example 12: Radiant-complexion gel**

FORMULA

	A	Composition 1:	4%
		Water:	30%
	B	Elastine HPM:	5.0%
20	C	Micropearl™ M 100:	3%
		Water:	5%
	D	Sepicide™ CL:	0.2%
		Sepicide™ HB:	0.3%
		Fragrance:	0.06%
25		50% sodium pyrrolidinone- carboxylate:	1%
		Water:	qs 100%

PROCEDURE

Prepare A; add B and then C, followed by D.

30

**Example 13: Body milk**

FORMULA

	A	Sepiperl™ N:	3.0%
		Glyceryl triheptonate:	10.0%
35	B	Water:	qs 100%
	C	Composition 1:	1.0%
	D	Fragrance:	qs
		Preserving agent:	qs

PROCEDURE

Melt A at about 75°C. Emulsify B in A at 75°C and then add C at about 60°C, followed by D.

5 **Example 14: Make-up-removing emulsion containing sweet almond oil**

FORMULA

	Montanov™ 68:	5%
	Sweet almond oil:	5%
10	Water:	qs 100%
	Composition 1:	0.3%
	Glycerol:	5%
	Preserving agent:	0.2%
	Fragrance:	0.3%

15

**Example 15: Moisturizing cream for greasy skin**

FORMULA

	Montanov™ 68:	5%
	Cetylstearyl octanoate:	8%
20	Octyl palmitate:	2%
	Water:	qs 100%
	Composition 1:	0.6%
	Micropearl™ M100:	3.0%
	Mucopolysaccharides:	5%
25	Sepicide™ HB:	0.8%
	Fragrance:	0.3%

**Example 16: Alcohol-free soothing aftershave balm**

FORMULA

30	Mixture of lauryl amino acids:	0.1% to 5%
	Magnesium potassium aspartate:	0.002% to 0.5%
	Lanol™ 99:	2%
	Sweet almond oil:	0.5%
	Water:	qs 100%
35	Composition 1:	3%
	Sepicide™ HB:	0.3%
	Sepicide™ CI:	0.2%
	Fragrance:	0.4%

**Example 17: Cream containing AHAs for sensitive skin**

FORMULA

	Mixture of lauryl amino acids:	0.1% to 5%
	Magnesium potassium aspartate:	0.002% to 0.5%
5	Lanol <sup>TM</sup> 99:	2%
	Montanov <sup>TM</sup> 68:	5.0%
	Water:	qs 100%
	Composition 1:	1.50%
	Gluconic acid:	1.50%
10	Triethylamine:	0.9%
	Sepicide <sup>TM</sup> HB:	0.3%
	Sepicide <sup>TM</sup> CI:	0.2%
	Fragrance:	0.4%

**Example 18: Aftersun soothing care**

FORMULA

	Mixture of lauryl amino acids:	0.1% to 5%
	Magnesium potassium aspartate:	0.002% to 0.5%
	Lanol <sup>TM</sup> 99:	10.0%
20	Water:	qs 100%
	Composition 1:	2.50%
	Sepicide <sup>TM</sup> HB:	0.3%
	Sepicide <sup>TM</sup> CI:	0.2%
	Fragrance:	0.4%
25	Colorant:	0.03%

**Example 19: Make-up-removing milk**

FORMULA

	Sepiperl <sup>TM</sup> N:	3%
30	Primol <sup>TM</sup> 352:	8.0%
	Sweet almond oil:	2%
	Water:	qs 100%
	Composition 1:	0.8%
	Preserving agent:	0.2%

35

**Example 20: Body milk**

FORMULA

	Sepiperl <sup>TM</sup> N:	3.5%
	Lanol <sup>TM</sup> 37T:	8.0%

	Solagum <sup>TM</sup> L:	0.05%
	Water:	qs 100%
	Benzophenone:	2.0%
	Dimethicone 350 cPs:	0.05%
5	Composition 1:	0.8%
	Preserving agent:	0.2%
	Fragrance:	0.4%

**Example 21: Fluid emulsion of alkaline pH**

10	Marcol <sup>TM</sup> 82:	5.0%
	NaOH:	10.0%
	Water:	qs 100%
	Composition 1:	1.5%

**Example 22: Fluid foundation**

FORMULA

	Simulsol <sup>TM</sup> 165:	5.0%
	Lanol <sup>TM</sup> 84D:	8.0%
	Lanol <sup>TM</sup> 99:	5.0%
20	Water:	qs 100%
	Mineral pigments and fillers:	10.0%
	Composition 1:	1.2%
	Preserving agent:	0.2%
	Fragrance:	0.4%

25

**Example 23: Antisun milk**

FORMULA

	Sepiperl <sup>TM</sup> N:	3.5%
	Lanol <sup>TM</sup> 37T:	10.0%
30	Parsol Nox <sup>TM</sup> :	5.0%
	Eusolex <sup>TM</sup> 4360:	2.0%
	Water:	qs 100%
	Composition 1:	1.8%
	Preserving agent:	0.2%
35	Fragrance:	0.4%



**Example 24: Eye contour gel**

FORMULA

	Composition 1:	2.0%
	Fragrance:	0.06%
5	Sodium pyrrolidinone- carboxylate:	0.2%
	Dow Corning™ 245 fluid:	2.0%
	Water:	qs 100%

10 **Example 25: Leave-on care composition**

FORMULA

	Composition 1:	1.5%
	Fragrance:	qs
	Preserving agent:	qs
15	Dow Corning™ X2 8360:	5.0%
	Dow Corning™ Q2 1401:	15%
	Water:	qs 100%

**Example 26: Slimming gel**

20	Composition 1:	5%
	Ethanol:	30%
	Menthol:	0.1%
	Caffeine:	2.5%
	Extract of ruscus:	2%
25	Extract of ivy:	2%
	Sepicide™ HP:	1%
	Water:	qs 100%

**Example 27: Alcohol-free soothing aftershave balm**

30 FORMULA

A	Lipacide™ PVB:	1.0%
	Lanol™ 99:	2.0%
	Sweet almond oil:	0.5%
B	Composition 1:	3.5%
35 C	Water:	qs 100%
D	Fragrance:	0.4%
	Sepicide™ HB:	0.4%
	Sepicide™ CI:	0.2%

**Example 28: Refreshing aftershave gel**

FORMULA

5	A	Lipacide™ PVB:	0.5%
		Lanol™ 99:	5.0%
		Composition 1:	2.5%
	B	Water:	qs 100%
10	C	Micropearl™ LM:	0.5%
		Fragrance:	0.2%
		Sepicide™ HB:	0.3%
		Sepicide™ CI:	0.2%

**Example 29: Care for greasy skin**

FORMULA

15	A	Micropearl™ M310:	1.0%
		Composition 1:	5.0%
		Octyl isononanoate:	4.0%
	B	Water:	qs 100%
20	C	Sepicontrol™ A5:	4.0%
		Fragrance:	0.1%
		Sepicide™ HB:	0.3%
		Sepicide™ CI:	0.2%
	D	Capigel™ 98:	0.5%
		Water:	10%

**Example 30: Cream containing AHAs**

FORMULA

30	A	Montanov™ 68:	5.0%
		Lipacide™ PVB:	1.05%
		Lanol™ 99:	10.0%
	B	Water:	qs 100%
		Gluconic acid:	1.5%
		TEA (triethylamine):	0.9%
	C	Composition 1:	1.5%
35	D	Fragrance:	0.4%
		Sepicide™ HB:	0.2%
		Sepicide™ CI:	0.4%

**Example 31: Nongreasy self-tanning agent for the face and body**

FORMULA

A	Lanol™ 2681:	3.0%
5	Composition 1:	2.5%
B	Water:	qs 100%
	Dihydroxyacetone:	3.0%
C	Fragrance:	0.2%
	Sepicide™ HB:	0.8%
10	NaOH (sodium hydroxide):	qs pH = 5%

**Example 32: Antisun milk containing Tahitian monoi**

FORMULA

A	Tahitian monoi:	10%
15	Lipacide™ PVB:	0.5%
	Composition 1:	2.2%
B	Water:	qs 100%
C	Fragrance:	0.1%
	Sepicide™ HB:	0.3%
20	Sepicide™ CI:	0.1%
	Octyl methoxycinnamate:	4.0%

**Example 33: Antisun care for the face**

FORMULA

25	A	Cyclomethicone and dimethiconol:	4.0%
		Composition 1:	3.5%
	B	Water:	qs 100%
	C	Fragrance:	0.1%
		Sepicide™ HB:	0.3%
30		Sepicide™ CI:	0.21%
		Octyl methoxycinnamate:	5.0%
		Titanium mica:	2.0%
		Lactic acid:	qs pH = 6.5

**Example 34: Self-tanning emulsion**

FORMULA

A	Lanol™ 99:	15%
	Montanov™ 68:	5.0%
	Octyl para-methoxycinnamate:	3.0%

	B	Water:	qs 100%
		Dihydroxyacetone:	5.0%
		Monosodium phosphate:	0.2%
	C	Composition 1:	0.5%
5	D	Fragrance:	0.3%
		Sepicide™ HB:	0.8%
		NaOH:	qs pH = 5

10 Montanov™ 68 (cetearyl glucoside) is a self-emulsifying composition as disclosed in WO 92/06778, sold by the company SEPPIC.

Micropearl™ M 100 is an ultrafine powder with a very soft feel and a matt effect, sold by the company Matsumo.

15 Sepicide™ CI, imidazolineurea, is a preserving agent sold by the company SEPPIC.

Pemulen™ TR is an acrylic polymer sold by Goodrich.

Simulsol™ 165 is self-emulsifying glyceryl stearate, sold by the company SEPPIC.

20 Lanol™ 1688 is a nongreasy emollient ester sold by the company SEPPIC.

Lanol™ 14M and Lanol® S are consistency factors sold by the company SEPPIC.

25 Sepicide™ HB, which is a mixture of phenoxyethanol, methyl paraben, ethyl paraben, propyl paraben and butyl paraben, is a preserving agent sold by the company SEPPIC.

Monteine™ CA is a moisturizer sold by the company SEPPIC.

30 Schercemol™ OP is a nongreasy emollient ester.

Lanol™ P is a stabilizing additive sold by the company SEPPIC.

Parsol™ MCX is octyl para-methoxycinnamate, sold by the company Givaudan.

35 Sepiperl™ N is a nacreous agent sold by the company SEPPIC, based on a mixture of alkylpolyglucosides such as those disclosed in WO 95/13863.

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Lanol™ 99 is isononyl isononanoate, sold by the company SEPPIC.

Solagum<sup>TM</sup> L is a carrageenan sold by the company SEPPIC.

Lanol™ 84D is dioctyl malate, sold by the company SEPPIC.

Eusolex<sup>TM</sup> 4360 is a sunscreen sold by the company Merck.

Lipacide™ PVB is a palmitoylated wheat protein hydrolyzate, sold by the company SEPPIC.

Sepicontrol™ A5 is a mixture of capryloylglycine, sarcosine and extract of *Cinnamomum zeylanicum*, sold by

Capigel™ 98 is an acrylate copolymer sold by the company SEPPIC.

Montanov<sup>TM</sup> 202 is a composition as disclosed in  
WO 98/47610, sold by the company SEPPIC.

**CLAIMS**

1. Process for preparing a composition in the form of an inverse latex, comprising an oil phase, an aqueous phase, at least one emulsifier of water-in-oil (W/O) type, at least one emulsifier of oil-in-water (O/W) type and from 20% to 75% by weight, mainly from 20% to 60% by weight and more particularly from 30% to 45% by weight, of a branched or vulcanized anionic polyelectrolyte, this process successively comprising:
- a step (a) of preparing an aqueous solution containing the monomers and the optional additives;
  - a step (b) of emulsifying the aqueous phase prepared in step (a), in an organic phase, in the presence of one or more emulsifiers of water-in-oil type;
  - a step (c) of polymerizing the monomers in the aqueous phase, initiated by introducing a free-radical initiator into said phase; and
  - a step (d) of adding one or more emulsifiers of oil-in-water type to the resulting dispersion, at a temperature of less than 50°C, characterized in that:
    - the polymerization reaction in step (c) is carried out at a pH of less than 5.5,
    - none of said emulsifiers belongs to the alkanolamide family,
    - the emulsifiers of the water-in-oil type used consist essentially of sorbitan monooleate or of sorbitan isostearate,
    - said anionic polyelectrolyte is based either on a monomer containing a strong acid function, or on at least one monomer containing a strong acid function copolymerized either with at least one monomer containing a weak acid function or with at least one neutral monomer, and

- from 30% to 80% and preferably 30% to 60%, in molar proportions, of the monomer moieties which the anionic polyelectrolyte comprises contain a strong acid function.

5 2. Process as defined in Claim 1, in which the reaction medium obtained from step (b) is concentrated by distillation before carrying out step (c).

3. Process as defined in either of Claims 1 and 2, in which the polymerization reaction is initiated with  
10 a redox couple which generates hydrogen sulfite ions ( $\text{HSO}_3^-$ ), such as the cumene hydroperoxide/sodium metabisulfite ( $\text{Na}_2\text{S}_2\text{O}_5$ ) couple or the cumene hydroperoxide/thionyl chloride ( $\text{SOCl}_2$ ) couple, at a temperature of less than or equal to  $10^\circ\text{C}$ , if desired,  
15 supplemented with a polymerization coinitiator, such as azobis(isobutyronitrile) (AIBN).

4. Process as defined in one of Claims 1 to 3, characterized in that the anionic polyelectrolyte comprises from 30% to 50% of a monomer comprising a  
20 strong acid function and from 70% to 50% either of a monomer comprising a weak acid function or of a neutral monomer.

5. Process as defined in Claim 4, characterized in that the anionic polyelectrolyte comprises, in molar proportions, from 30% to 50% of 2-methyl-2-[(1-oxo-2-propenyl)amino]-1-propanesulfonic [lacuna], partially  
25 or totally salified in the form of an alkali metal salt, preferably the sodium salt, or the ammonium salt, and from 70% to 50% either of acrylamide.

6. Process as defined in one of Claims 1 to 5, characterized in that the anionic polyelectrolyte is vulcanized and/or branched with a diethylenic or polyethylenic compound in a molar proportion, expressed  
30 relative to the monomers used, of from 0.005% to 1% and preferably from 0.01% to 0.1%.

7. Process as defined in Claim 6, characterized in that the vulcanizing agent and/or branching agent is

chosen from ethylene glycol methacrylate, sodium diallyloxyacetate, ethylene glycol diacrylate, diallylurea, trimethylolpropane triacrylate and, more particularly, methylenebis(acrylamide).

5 8. Process as defined in one of Claims 1 to 7, characterized in that the oil phase represents from 15% to 40% and preferably from 20% to 25% of its total weight.

10 9. Process as defined in one of Claims 1 to 8, characterized in that the oil phase consists essentially of isohexadecane or of white mineral oil.

15 10. Use of the composition obtained according to the process as defined in one of Claims 1 to 9, to prepare a cosmetic, dermopharmaceutical or pharmaceutical topical composition.

11. Cosmetic, dermopharmaceutical or pharmaceutical composition comprising from 0.1% to 10% by weight of an inverse latex obtained according to the process as defined in one of Claims 1 to 9.

20 12. Composition as defined in Claim 11, in the form of a milk, a lotion, a gel, a cream, a soap, a bubble bath, a balm, a shampoo or a conditioner.



## ABSTRACT

Process for preparing a composition in the form of an inverse latex, comprising an oil phase, an aqueous phase, at least one emulsifier of water-in-oil (W/O) type, at least one emulsifier of oil-in-water (O/W) type and from 20% to 75% by weight, mainly from 20% to 60% by weight and more particularly from 30% to 45% by weight, of a branched or vulcanized anionic polyelectrolyte, characterized in that the polymerization reaction is carried out at a pH of less than 5.5, in that the emulsifiers do not belong to the alkanolamide family and in that said anionic polyelectrolyte is based either on a monomer containing a strong acid function, or on at least one monomer containing a strong acid function copolymerized either with at least one monomer containing a weak acid function or with at least one neutral monomer. Compositions obtained and cosmetic applications.

0953903-090701

# COMBINED DECLARATION AND POWER OF ATTORNEY

As a below named inventor, I hereby declare that

My residence, post office address and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

## NOVEL ALKANOLAMIDE-FREE THICKENING LATEX

the specification of which: *(check one)*

### REGULAR OR DESIGN APPLICATION

☐ is attached hereto.

☐ was filed on \_\_\_\_\_ as application Serial No. \_\_\_\_\_ and was amended on (if applicable).

### PCT FILED APPLICATION ENTERING NATIONAL STAGE

☒ was described and claimed in International application PCT/FR99/02886 filed on 23 November 1999 and as amended on (if any).

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, §1.56.

### PRIORITY CLAIM

I hereby claim foreign priority benefits under 35 USC 119 of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed.

### PRIOR FOREIGN APPLICATION(S)

Country	Application Number	Date of Filing (day, month, year)	Priority Claimed
France	98 14965	27 November 1998	yes

*(Complete this part only if this is a continuing application.)*

I hereby claim the benefit under 35 USC 120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of 35 USC 112, I acknowledge the duty to disclose information which is material to patentability as defined in Title 37 Code of Federal Regulations §1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application:

(Application Serial No.)

(Filing Date)

(Status--patented, pending, abandoned)

# POWER OF ATTORNEY

The undersigned hereby authorizes the U.S. attorney or agent named herein to accept and follow instructions from L'Air Liquide as to any action to be taken in the Patent and Trademark Office regarding this application without direct communication between the U.S. attorney or agent and the undersigned. In the event of a change in the persons from whom instructions may be taken, the U.S. attorney or agent named herein will be so notified by the undersigned.

As a named inventor, I hereby appoint the registered patent attorneys represented by Customer No. **000466** to prosecute this application and transact all business in the Patent and Trademark Office connected therewith, including: **Robert J. PATCH, Reg. No. 17,355, Andrew J. PATCH, Reg. No. 32,925, Robert F. HARGEST, Reg. No. 25,590, Benoît CASTEL, Reg. No. 35,041, Eric JENSEN, Reg. No. 37,855, Thomas W. PERKINS, Reg. No. 33,027, and Roland E. LONG, Jr., Reg. No. 41,949,**

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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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